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Lead Zirconate Titanate Thick-Film Ultrasonic Transducer for 1 to 20 MHz Frequency Bands Fabricated by Hydrothermal Polycrystal Growth

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A thickness vibration mode ultrasonic transducer was fabricated by hydrothermal deposition of lead zirconate titanate polycrystalline thick film. This hydrothermal polycrystalline PZT thick-film (HPTF-PZT) ultrasonic transducer was studied for its wide-frequency-band transmission characteristics. The transducer had a 50-μm-thick PZT layer on a 50-μm-thick titanium substrate in an active area of 8 mm × 8 mm. The experimental results showed the HPTF-PZT ultrasonic transducer radiated a single ultrasound pulse in the frequency range from 1 to 20 MHz with a 20 dB deviation. Additionally, this transducer generated a 2 MHz sawtooth ultrasonic wave with odd-and even order-harmonic components from the 2nd to the 11th harmonics. These results indicate that the fabricated ultrasonic transducer has satisfactory wide-band characteristics for a MHz frequency range ultrasonic transmitter. [DOI: 10.1143/JJAP.44.4342]

KEYWORDS: hydrothermal method, PZT thick film, microstructure, low acoustic impedance material, ultrasonic transducer, wide-band characteristics

1. Introduction

Ultrasonic transducers with a frequency range from 1 to 50 MHz have been widely utilized for medical ultrasonic imaging and nondestructive evaluation. These ultrasonic transducers are expected to have wide-band characteristics. Because a wide-band ultrasonic wave is beneficial to high-quality imaging, graded piezoelectric plates[1)] and single crystals of high mechanical coupling factor and low-acoustic impedance materials[2,3)] have been proposed for use in wide/broad-band ultrasonic transducers. However, it is difficult to fabricate wide-band ultrasonic transducers operating above 10 MHz.

In this study, a novel wide-band ultrasonic transducer operating in the range of 1 to 20 MHz using a hydrothermal PZT thick film is proposed. The hydrothermal method is known as a fabrication technique for a variety of crystals, powders and ingots. However, in this study the hydrothermal method is used to deposit polycrystalline PZT on Ti substrates.[4–6)] Additionally, this hydrothermal PZT thick film has many favorable features as follows: the film may be deposited on concave or convex Ti substrates at a thickness of 10 μm over a thick PZT film; the boundary of the Ti substrate and PZT films dose not easily peel, poling and annealing are not required; and the piezoelectric transverse effect causes a high vibration velocity of its bending[5,6)] and longitudinal modes.[7)] Because of its characteristics, this material seems to be a good ultrasonic transducer for transmission and reception at 10 MHz and over. A transducer was fabricated by depositing a 50 μm PZT thick film on 50-μm-thick Ti substrate using the hydrothermal method. The thickness mode resonance frequency of this HPTF-PZT transducer was estimated to be between 10 MHz and 20 MHz. Its wide-band transmission characteristics as an ultrasonic transducer were investigated.

2. Hydrothermal Method for PZT Deposition

The hydrothermal method was used to deposit PZT crystals on a Ti substrate. First, PZT nuclei were deposited on a Ti substrate from a solution using an autoclave, and then the crystals were grown to the target thickness, respectively. The temperature and pressure under deposition conditions were 180°C and 0.95 MPa. The reaction time was 12 h for each process. Starting materials for the reaction were lead nitrate [$Pb(NO_3)_2$ 99.0%], zirconium chloride oxide octahydrate ($ZrOCl_2 \cdot 8\ H_2O$ 99.0%), and a powder of titanium oxide (TiO_2 99.0%) in potassium hydroxide (KOH) solution.[8)] The composition of the starting materials was determined as follows: 87 ml of 1 N aqueous $Pb(NO_3)_2$, 52 ml of 1 N aqueous $ZrOCl_2 \cdot 8\ H_2O$, 1 g TiO_2, and 200 ml of 4 N KOH solution. The solution filled about 60% of the autoclave capacity. The Ti substrates were fixed on stirrer plates made of Teflon, and the stirrers were kept in the solution. The stirrers were rotated by a motor at 245 rpm; the tangential velocity of the substrate was about 0.8 m/s. The HPTF-PZT fabricated was about 50 μm thick as a consequence of running the process 20 times.

3. Deposited Film and Resonance Frequency Estimation

The HPTF-PZT morphology and microstructure were investigated by scanning electron microscopy (SEM). The phase structure of the films was evaluated by X-ray diffraction (XRD) and energy dispersion X-ray spectrometry (EDS). The HPTF-PZT had a polycrystalline structure, as shown in Fig. 1. It can be seen that the surface and the inside were ridged due to disordered crystal growth. Therefore, the density and elastic constant of the HPTF-PZT was about 50% and 30%, respectively of sintered ceramic PZT. However, the XRD pattern indicated a good crystal structure of PZT, as shown in Fig. 2. In addition, the atomic ratio of Zr : Ti in HPTF-PZT was investigated by EDS. The atomic ratio of the film was Zr : Ti = 80 : 20 (error ratio ±4%).

The acoustic velocity and acoustic impedance of HPTF-PZT were estimated using the reported value of density[4)] and elastic modulus.[8)] The phase velocity of a dilatational wave was 2200 m/s[8)] and the acoustic impedance was 8.6×10^6 Ns/m^3.[8)] The HPTF-PZT was identified as a low acoustic impedance material. The mechanical resonance frequency of the thickness mode is approximated[9)] using

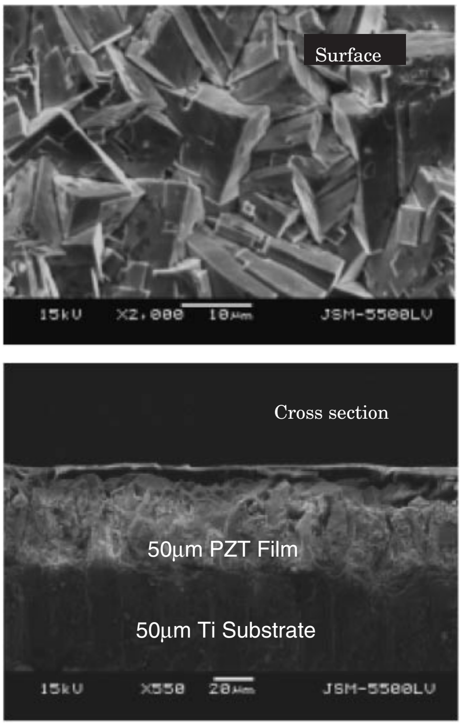

Fig. 1. SEM photographs of surface and thickness of hydrothermal PZT thick films.

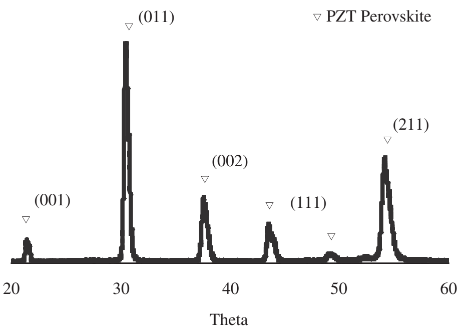

Fig. 2. XRD pattern of hydrothermal PZT films.

$$f_{\mathrm{pzt1/2}} = \frac{V_{\mathrm{pzt}}}{2t_{\mathrm{pzt}}}, \tag{1}$$

$$f_{\mathrm{pzt1/4}} = \frac{V_{\mathrm{pzt}}}{4t_{\mathrm{pzt}}}, \tag{2}$$

$$f_{\mathrm{pzt1+ti}} = \frac{1}{2\left(\dfrac{t_{\mathrm{pzt}}}{V_{\mathrm{pzt}}} + \dfrac{t_{\mathrm{ti}}}{V_{\mathrm{ti}}}\right)}, \tag{3}$$

where $f_{\mathrm{pzt1/2}}$ is the half wavelength resonance frequency of the PZT film, $f_{\mathrm{pzt1/4}}$ is the quarter wavelength resonance frequency of the PZT film, and $f_{\mathrm{pzt+ti}}$ is the half wavelength resonance frequency of the vibration plate of the PZT film-Ti substrate. t_{pzt} and t_{ti} are the thicknesses of the materials, and V_{pzt} and V_{ti} are the phase velocities of the dilatational waves. $f_{\mathrm{pzt1/2}}$ is 26 MHz, the $f_{\mathrm{pzt1/4}}$ is 13 MHz, and $f_{\mathrm{pzt+ti}}$ is 18 MHz.

4. Transducer Fabrication

The HPTF-PZT was deposited on a 12 mm × 12 mm × 50 μm Ti substrate that was used as an electrode, and the counter electrode was an 8 mm × 8 mm Au square on the surface of the PZT film. Au was deposited by vacuum evaporation. A lead wire was attached to the Ti substrate and the Au electrode. The HPTF-PZT had the same direction of spontaneous polarization during the hydrothermal process. Therefore, the HPTF-PZT transducer was nonpoling. The transducer had no structures for backing and matching layers. The transducer was fixed at its edges to keep the degassed water inside.

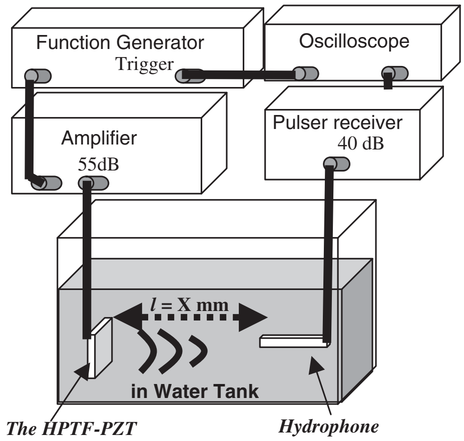

Fig. 3. Schematic diagram of measurement setup for ultrasound transmission.

5. Experiments for Ultrasound Generation

The transmission characteristics of the HPTF-PZT ultrasonic transducer were investigated by measuring the frequency response of ultrasound transmission and higher harmonic frequency generation in water. The experimental setup is shown in Fig. 3. The ultrasonic transducer was driven by a signal from a function generator. The signal was amplified by an amplifier (ENI A300) with a 55 dB gain factor. The generated ultrasound propagated in the water. The ultrasound was detected by a needle-type hydrophone. The detected wave signal was amplified by a pulser receiver at 40 dB, and this signal was displayed on an oscilloscope. The hydrophone (MHA500A) was calibrated in sensitivity from 1 to 20 MHz, as shown in Fig. 4. The pulser receiver had a high-pass filter at 1 MHz and a low-pass filter at 50 MHz. In addition, the Fresnel region[10] of this HPTF-PZT ultrasonic transducer were approximately 70 mm at 10 MHz and 136 mm at 20 MHz.

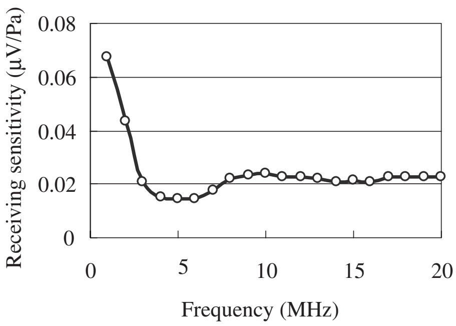

Fig. 4. Frequency characteristic of receiving sensitivity of utilized hydrophone.

6. Experimental Results of Frequency Response

The frequency response of the transducer was measured using a single 41 ns pulse whose amplitude was 25 V, as shown in Fig. 5. The transmission characteristics of the HPTF-PZT transducer were investigated using the impulse response in the MHz frequency band. The ultrasound radiating from the HPTF-PZT ultrasonic transducer was detected by the needle-type hydrophone at distances of 30 mm, 40 mm and 50 mm. The detected ultrasound waveforms are shown in Fig. 6 under the premise that the wave propagated directly between the transducer and the hydrophone in water. The propagation times were approximately 20 µs, 27 µs and 34 µs at distances of 30 mm, 40 mm and 50 mm, respectively; the reflected-wave traveling distance was maintained at more than 10 cm to avoid the appearance of this wave among the detected signals, as indicated in Fig. 6. The sound pressure was estimated using the receiving sensitivity of the hydrophone at 20 MHz. The sound pressures were 11 kPa, 29 kPa and 21 kPa at the distances of 30 mm, 40 mm and 50 mm, respectively.

The experimental results of the frequency response are shown in Figs. 7–9 at distances of 30 mm, 40 mm and 50 mm. In Figs. 7–9, (a) and (b) indicate the wave forms received by the hydrophone and the power spectra of the detected waves respectively. These power spectra were compensated by the frequency characteristics of the driving signal, as shown in Fig. 5. From Figs. 7(b)–9(b), it is found that the HPTF-PZT transducer had two broad mechanical resonances approximately 10 MHz and 20 MHz. Because of

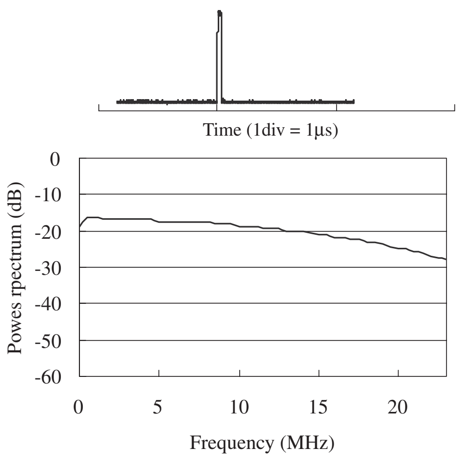

Fig. 5. (a) Driving signal and (b) frequency spectrum at a single 41 ns pulse.

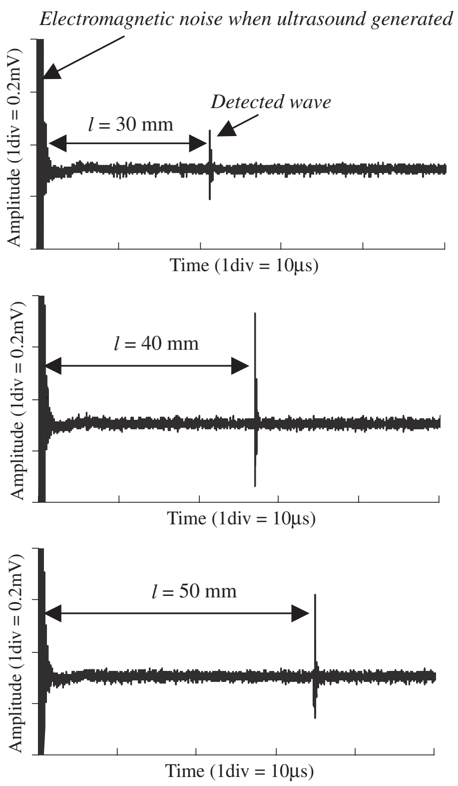

Fig. 6. Ultrasonic single pulses detected by hydrophone that radiated from HTPT-PZT ultrasonic transducer.

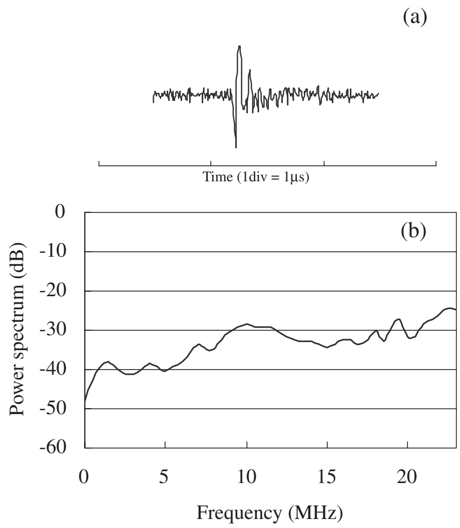

Fig. 7. (a) Waveform and (b) frequency spectrum at 41 ns single-cycle pulse generation at distance of 30 mm determined using HPTF-PZT ultrasonic transducer.

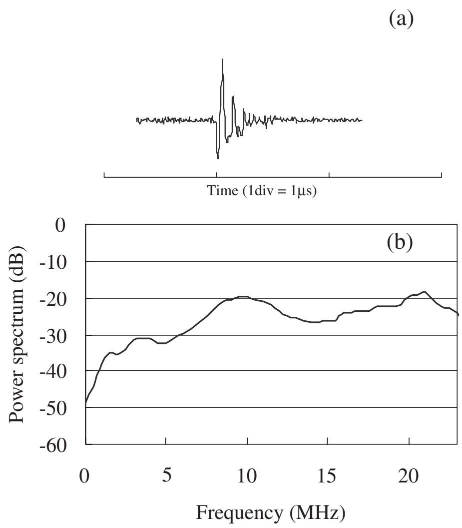

Fig. 8. (a) Waveform and (b) frequency spectrum at 41 ns single-cycle pulse generation at distance of 40 mm determined using HPTF-PZT ultrasonic transducer.

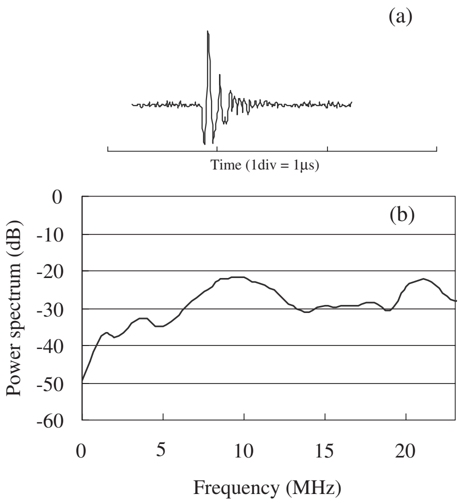

Fig. 9. (a) Waveform and (b) frequency spectrum at 41 ns single-cycle pulse generation at distance of 50 mm determined using HPTF-PZT ultrasonic transducer.

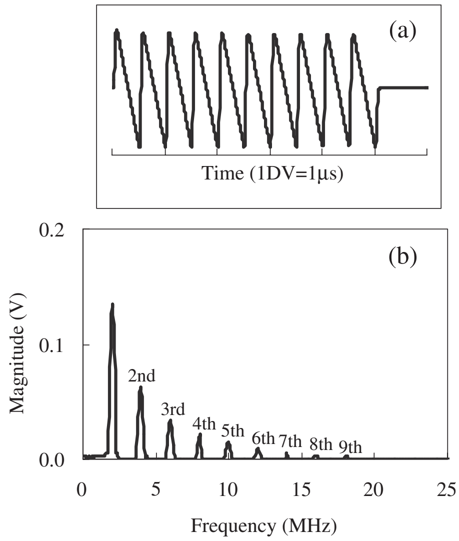

Fig. 10. (a) driving signal and (b) frequency spectrum of 2 MHz, 10-cycle sawtooth wave composed of odd-and even-order harmonics, and (c) generated waveform and (d) frequency spectrum determined using HPTF-PZT ultrasonic transducer at distance of 30 mm.

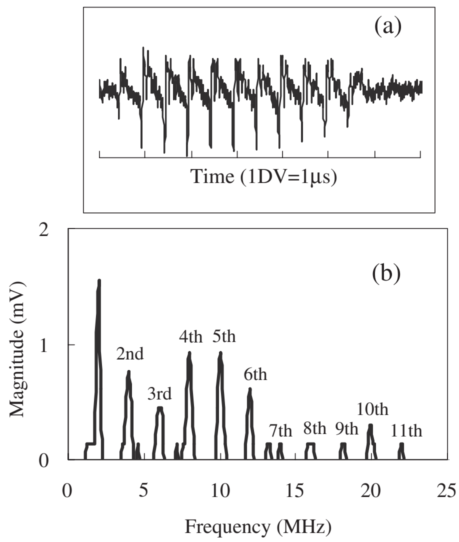

Fig. 11. (a) generated waveform and (b) frequency spectrum determined using HPTF-PZT ultrasonic transducer at distance of 30 mm.

these two broad resonances, the transducer had a wide frequency bandwidth from 1 to 20 MHz with a 20 dB deviation as indicated in the power spectra shown in Figs. 7–9. These results indicate that the HPTF-PZT transducer has wide-band characteristics. In addition, the propagated waves at distances of 30 mm, 40 mm and 50 mm are affected by the effect of the Fresnel region as mentioned previously. Hence, the frequency responses shown in Figs. 7–9 are slightly different from each other in terms of the amount of frequency components. However, the HPTF-PZT transducer had wide-band characteristics in all results.

7. Harmonic Generation Characteristics

The higher harmonic frequency generation of ultrasound was estimated. The driving signal was a 2 MHz, 10-cycle, 130 V sawtooth wave including "odd-and even-order harmonics" from the 2nd to the 11th order. The sawtooth wave drive experiment produced the harmonic generation of ultrasound. Figures 10(a) and 10(b) show the driving waveform and a spectrum of the driving waveform, respectively. Figures 11(a) and 11(b) show the generated waveform and a spectrum of the generated waveform, respectively. Figure 10(a) shows that the generated waveform rises steeply and drops off at the rise and fall of variations in sound pressure, respectively. It was almost a sawtooth wave at 2 MHz with the 2nd to 11th harmonics. Additionally, Fig. 11(b) shows the spectrum of the generated waveform excited at 10 MHz and 20 MHz in comparison with the driving signal. These results correspond to the estimated mechanical resonance frequency.

8. Conclusions

A novel 1 to 20 MHz wide-band ultrasonic transducer using HPTF-PZT was fabricated. The HPTF-PZT ultrasonic transducer was studied to determine its ultrasound transmission characteristics. It was shown that the HPF-PZT transducer had wide-frequency-band characteristics. From the impulse response, the transducer indicated a bandwidth from 1 to 20 MHz with a 20 dB deviation when the sound pressure was 29 kPa at a distance of 40 mm. This was a high-intensity sound pressure level for ultrasound transmission. The rectangular wave drive at 2 MHz generated higher odd-and even-order harmonic waves up to the 11th order, namely, a 22 MHz wave. Performance characteristics of the HPTF-PZT transducer above 20 MHz will be studied in the near future.

Acknowledgement

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